General Synthesis of Pd-pm (pm=Ga, In, Sn, Pb, Bi) Alloy Nanosheet Assemblies for Advanced Electrocatalysis

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Chemicals and Materials

Potassium tetrachloropalladate (K_2PdCl_4, 98%), indium chloride ($InCl_3$, 99%), gallium chloride ($GaCl_3$, 99.9%), bismuth acetate ($Bi(Ac)_3$, 99.8%), lead acetate ($Pb(Ac)_2$, 95%), sannous acetate ($Sn(Ac)_4$, 99.5%) ascorbic acid (AA, 99%), polyvinylpyrrolidone (PVP, MW: 8000), hexacarbonyl tungsten ($W(CO)_6$, 99%), N, N-Dimethylformamide (DMF, 99.5%), acetic acid (99.5%), ethylene glycol (EG, 98%). All other chemical reagents used in this work were analytically pure and used without further purification. All other chemical reagents used in this work were analytically pure and used without further purification.

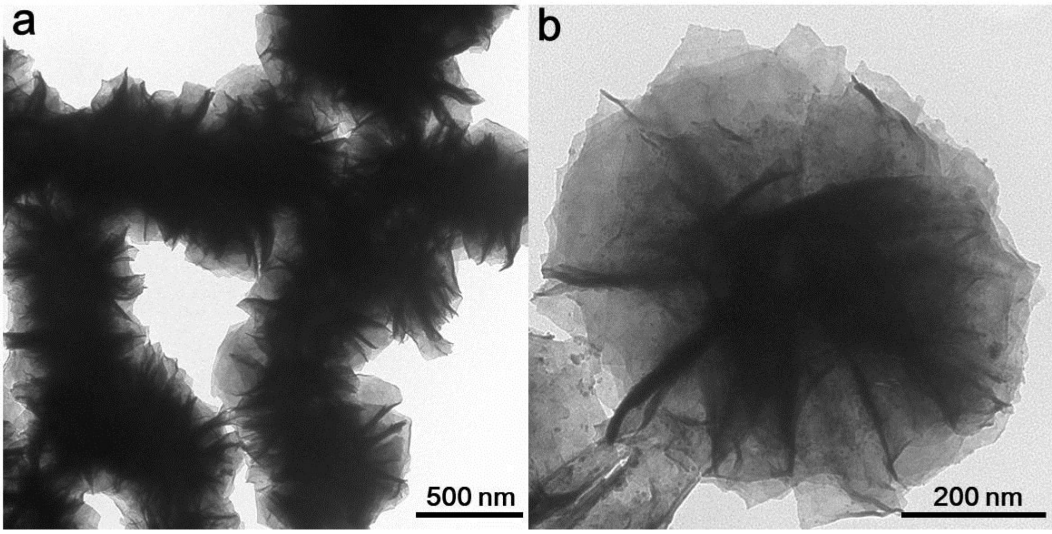

Fig.S1 TEM images of 3D flower-like Pd NSAs with different magnifications.

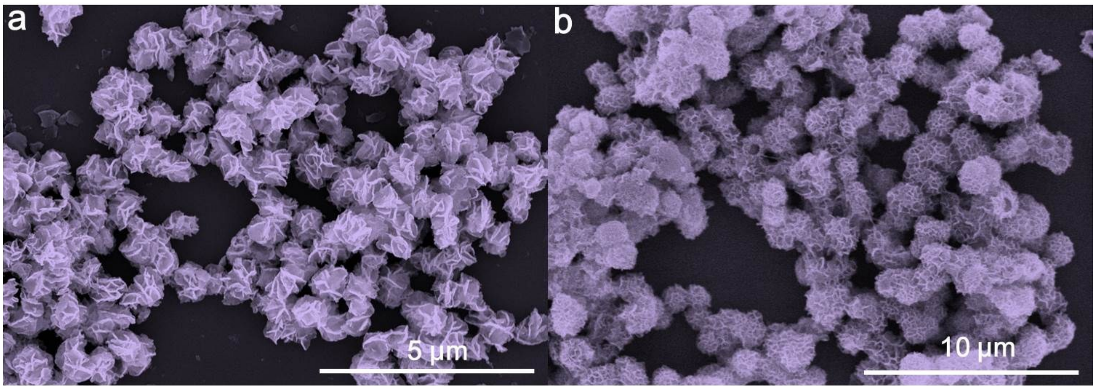

Fig.S2 Additional SEM images of (a) PdGa NSAs and (b) PdBi NSAs.

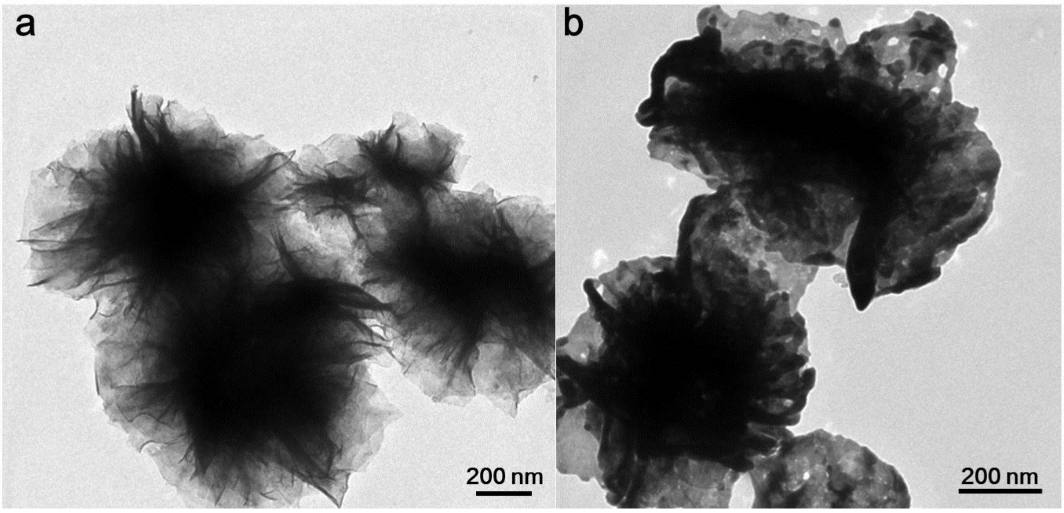

Fig.S3 Additional TEM images of (a) PdGa NSAs and (b) PdBi NSAs.

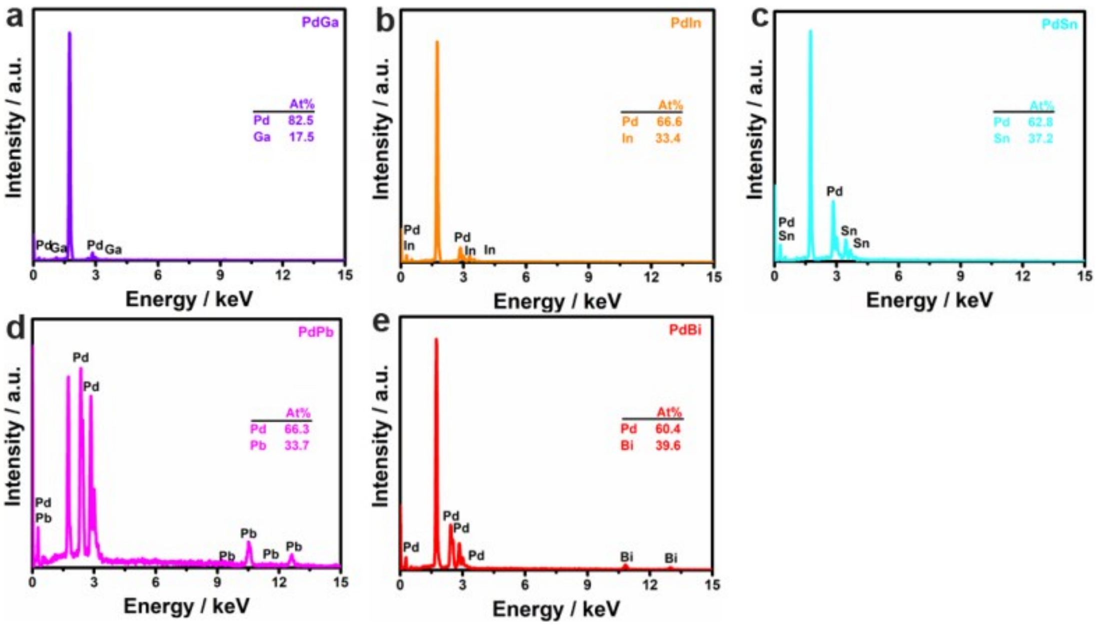

Fig.S4 SEM-EDX spectra of (a) PdGa NSAs, (b) PdIn NSAs, (c) PdSn NSAs, (d) PdPb NSAs, and (e) PdBi NSAs.

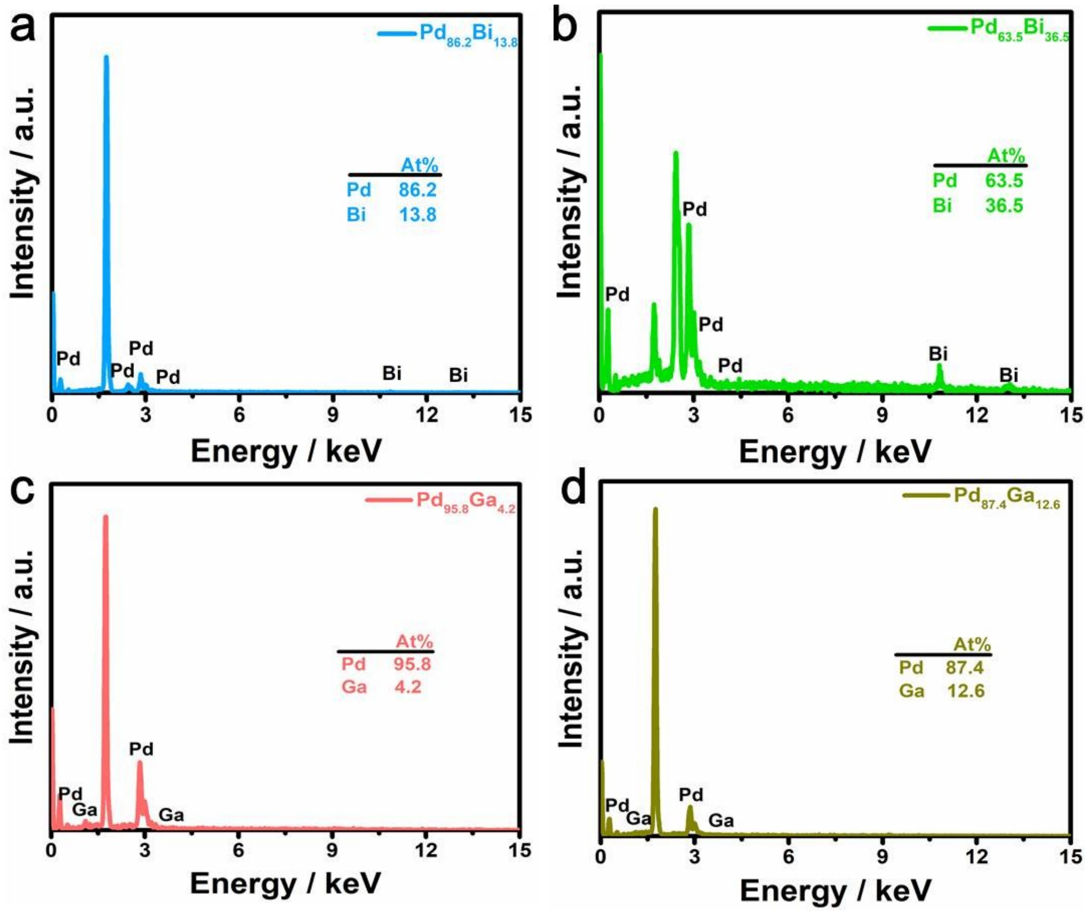

Fig.S5 SEM-EDX spectra of (a) $Pd_{86.2}Bi_{13.8}$, (b) $Pd_{63.5}Bi_{36.5}$, (c) $Pd_{95.8}Ga_{4.2}$, and (d) $Pd_{87.4}Ga_{12.6}$.

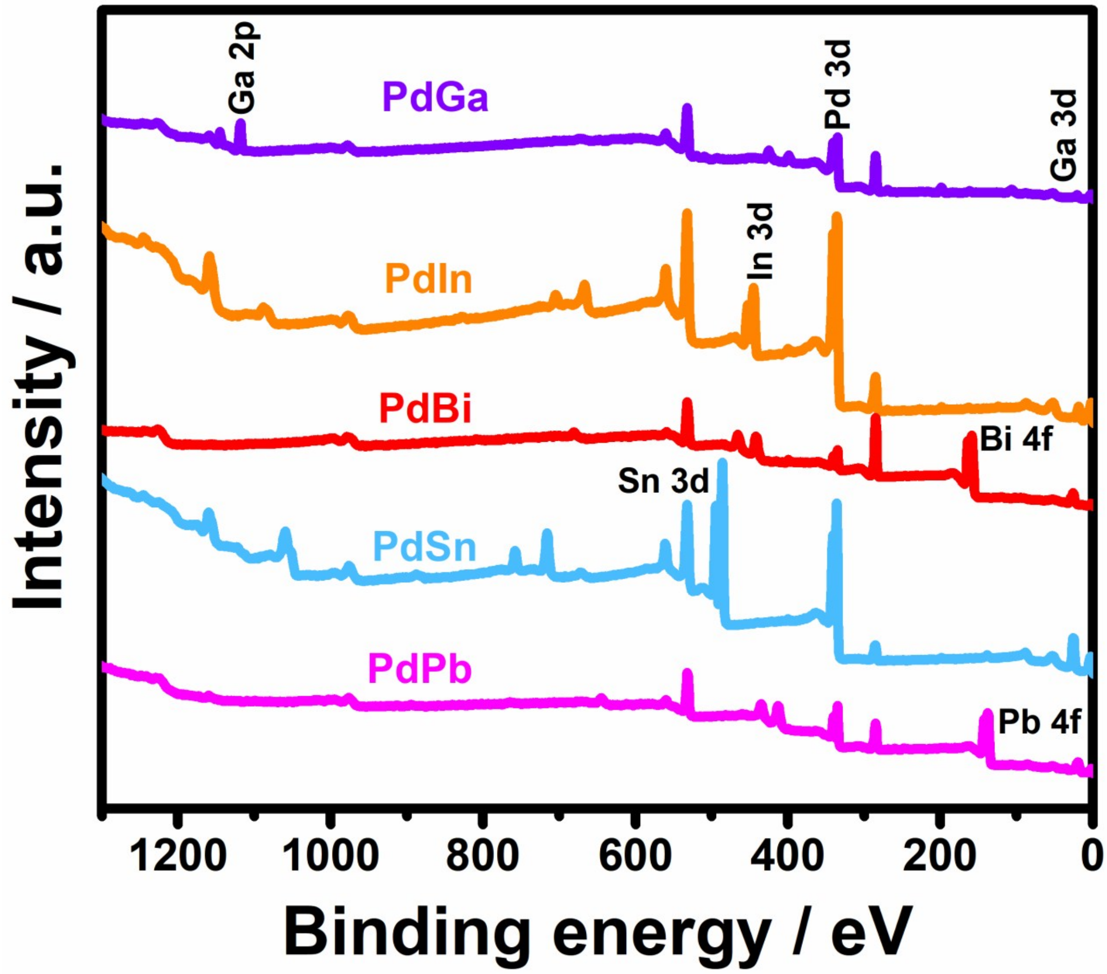

Fig.S6 Survey XPS spectra of PdGa NSAs, PdIn NSAs, PdBi NSAs, PdSn NSAs, and PdPb NSAs

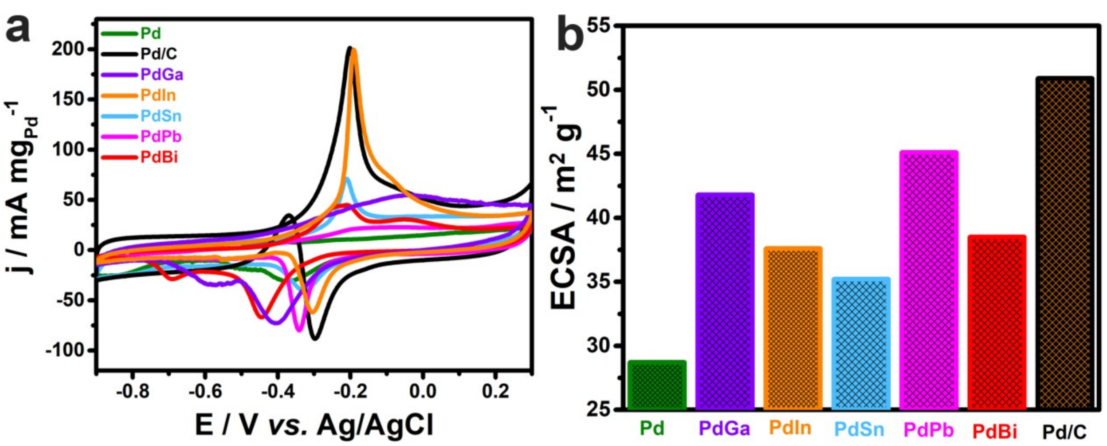

Fig.S7 (a) CV curves of Pd NSAs, Pd/C, PdGa NSAs, PdIn NSAs, PdBi NSAs, PdSn NSAs, and PdPb NSAs in 1 M KOH solution. (b) The histogram of the ECSA values for different catalysts.

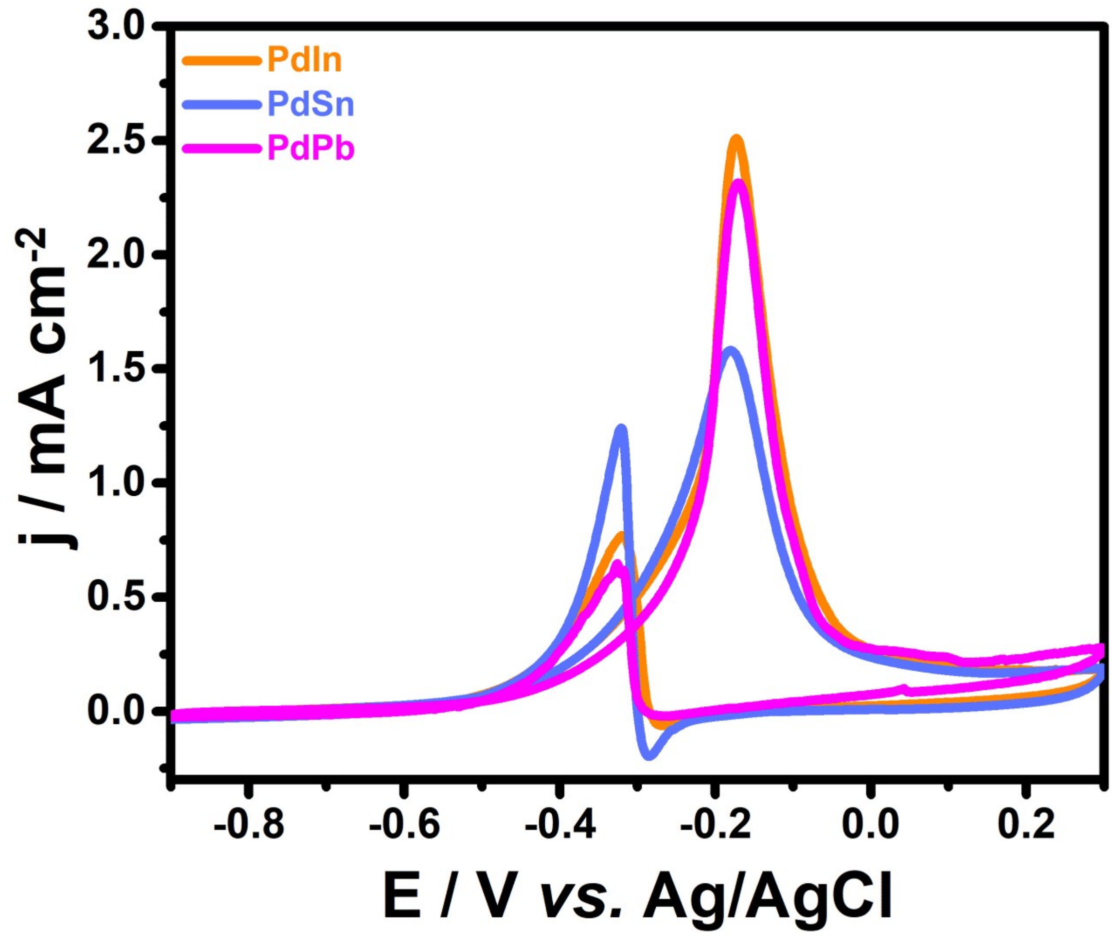

Fig.S8 ECSA-normalized CV curves of PdIn NSAs, PdSn NSAs, and PdSn NSAs in 1 M KOH + 1 M CH_3OH solution.

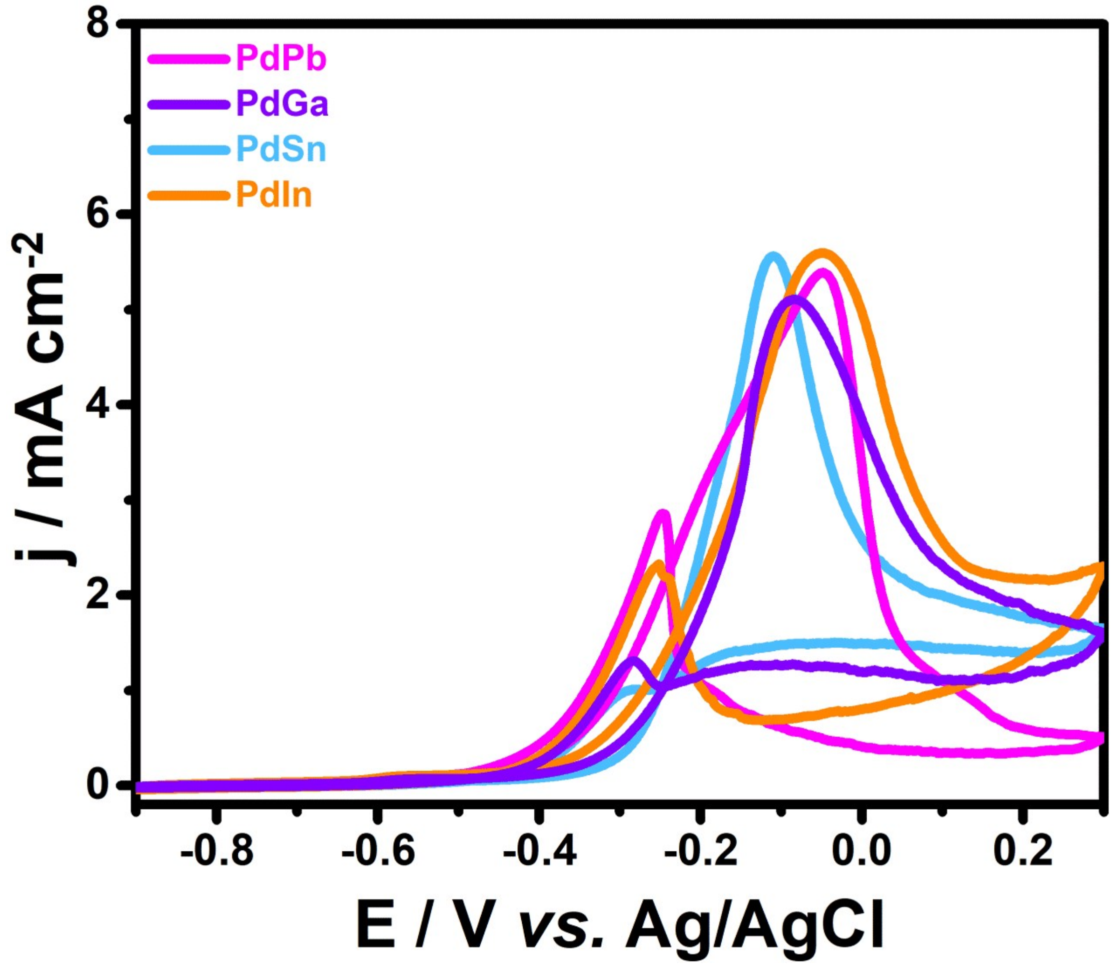

Fig.S9 ECSA-normalized CV curves of PdPb NSAs, PdIn NSAs, PdSn NSAs, and PdGa NSAs in 1 M KOH + 1 M glycerol solution.

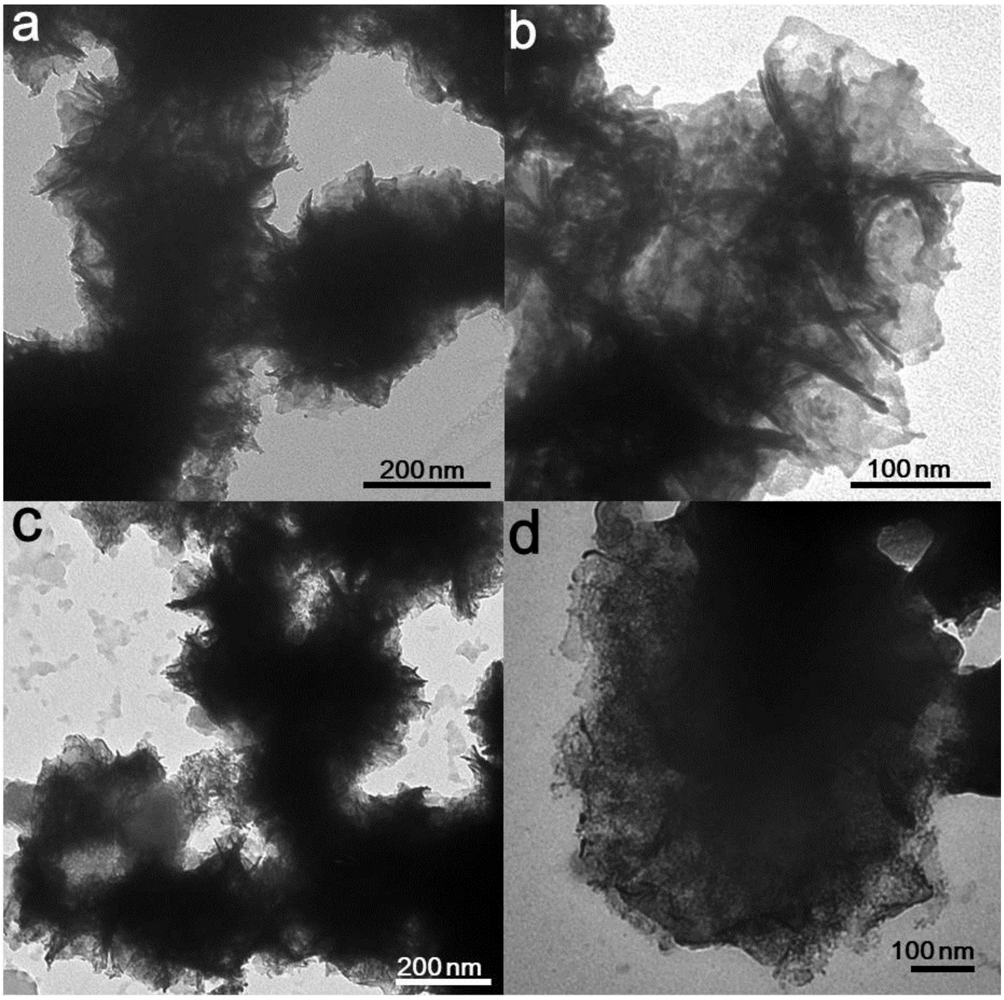

Fig.S10 Representative TEM images of (a and b) PdGa and (c and d) PdBi after electrochemical tests.

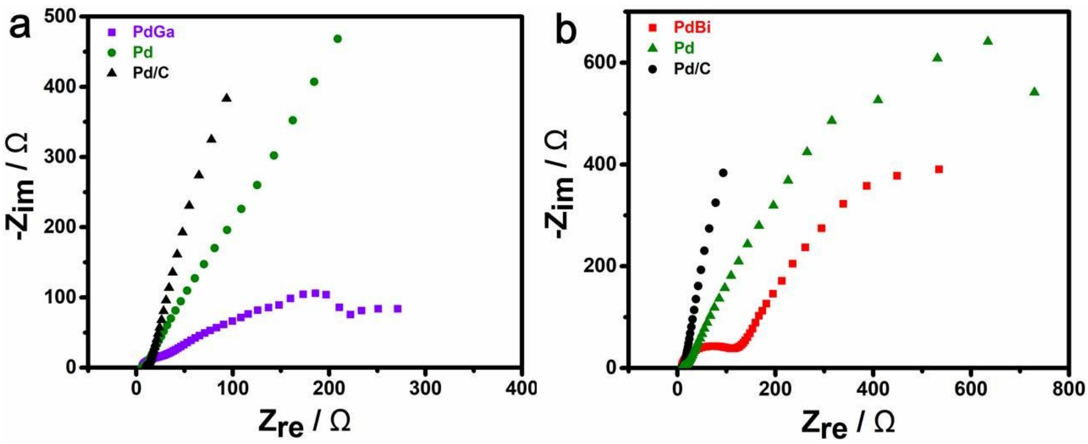

Fig.S11 Nyquist plots (potential= 0.2 V) of PdGa, Pd, and Pd/C in 1 M KOH + 1 M CH_3OH solution. Nyquist plots (potential= 0.2 V) of PdBi, Pd, and Pd/C in 1 M KOH + 1 M glycerol solution.

Table S1 MOR electrocatalytic activity comparison of PdGa NSAs with recently reported catalysts

Catalysts	Peaks currents from CV curves		Electrolyte	References
	J_m (A/mg)	J_s (mA/cm^2)		
PdGa NSAs	**1.1**	**2.65**	**1.0 M KOH + 1.0 M CH_3OH**	**This work**
Pd_2Cu_2/rGO	0.90		1 M KOH + 1 M CH_3OH	1
PdNi/RGO	0.80		0.5 M KOH + 1 M CH_3OH	2
Pd/P	0.844		1 M KOH + 1 M CH_3OH	3
Cu@Pt/C	0.62		0.5 M H_2SO_4 + 0.5 M CH_3OH	4
PdCuCo/RGO	1.06	~ 0.92	1 M KOH + 1 M CH_3OH	5
Pd NFs/PPy@MWCNTs	0.725	1.69	0.5 M KOH + 1 M CH_3OH	6
PtRu NWs	0.82	1.16	0.1 M $HClO_4$ + 0.5 M CH_3OH	7
Pt_3Cu/C	~ 0.70	~ 0.50	0.5 M $HClO_4$+ 1 M CH_3OH	8

Table S2 GOR electrocatalytic activity comparison of PdBi NSAs with recently reported catalysts

Catalysts	Peaks currents from CV curves		Electrolyte	References
	J_m (A/mg_{Pd})	J_s (mA/cm^2)		
PdBi NSAs	**3.04**	**7.9**	**1.0 M KOH + 1.0 M Glycerol**	**This work**
Pt flowers	0.18	0.32	0.5 M H_2SO_4 + 1 M Glycerol	9
$PtNi_{0.67}Pb_{0.26}$ NWs/C	0.36	0.61	0.1 M $HClO_4$ + 0.2 M Glycerol	10
Pd_5Ru-PEDOT/C		4.3	1 M KOH + 1 M Glycerol	11
$Pd_{50}Ni_{50}$/C	0.19		0.1 M KOH + 0.1 M Glycerol	12
Pd_3Sn/phen-C	0.175		0.1 M KOH + 0.5 M Glycerol	13
Pd-NiO_x-P/C	0.364		0.1 M KOH + 0.5 M Glycerol	14
$PdCu_2$	1.6		1 M KOH + 1 M Glycerol	15

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